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54 **Wettable olefin polymer fibers.**

57 Wettable fibers and/or filaments are prepared from olefin polymers, especially linear low density copolymer (LLDPE), by incorporating into said olefin polymer at least one wetting agent of the group comprising (a) an alkyoxylated alkyl phenol along with a mixed mono-, di-, and/or tri-glyceride, (b) a polyoxalkylene fatty acid ester, or (c) a combination of (b) and any or all parts of (a).

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## WETTABLE OLEFIN POLYMER FIBERS

Many olefin polymers can be made into fibers and filaments. This includes polyethylene, polypropylene, polybutene, polypentene, and ethylene copolymerized with  
5 other olefinic monomers such as higher olefins. Such olefin polymers are known for their hydrophobic properties. Wettability of the polymers, including those in fibrous or filament form, is achieved by means of wetting agents provided in or on the polymers.

10 U.S. 3,847,676, U.S. 4,073,852, U.S. 4,307,143, U.S. 4,273,892, and U.S. 4,274,971 are representative of this art. Also U.S. Patent 4,189,420 discloses certain ethylene polymers blended with a polybutene and a mixed glyceride having at least one acyl group of 2  
15 to 6 carbon atoms and at least one acyl group containing 8 to 22 carbon atoms. U.S. Patent 3,048,266 discloses an anti-fog agent of polyethylene oxide derivative in a polyolefin composition. U.S. Patent 3,048,263 discloses a polyolefin anti-fog agent comprising a monoglyceride  
20 of a fatty acid. U.S. Patent 2,462,331 discloses the incorporation into polyethylene of polyhydric alcohol esters or metal salts of either saturated or unsaturated monocarboxylic fatty acids.

The present invention pertains to an olefin polymer, especially a linear low density polyethylene copolymer (LLDPE), having compounded therewith a wetting agent, for use in forming wettable fibers and/or fine filaments. The wetting agent comprises at least one of the following: (1) an alkoxyated alkylphenol along with a mixed mono-, di-and/or tri-glyceride, (2) a polyoxalkylene fatty acid ester, or (3) a combination of (2) and any part of (1) such as polyalkylene fatty acid ester and an alkoxyated alkylphenol, a mixed mono-, di- and/or triglyceride, or both alkoxyated alkylphenol and mixed glyceride. More particularly it concerns wettable fibers or fine filaments prepared from linear low density polyethylene (LLDPE) and blends or alloys of LLDPE with other olefin polymers. Preferably such wettable fibers and fine filaments are extruded strands of less than about 15 denier.

#### DESCRIPTION

There are basically two polymerization techniques for preparing high molecular weight olefin polymers and copolymers. The oldest commercial technique involves high pressure, high temperature, and the use of a free radical initiator, such as a peroxide to give polymers generally known as low density polyethylene (LDPE). They are also known as ICI-type polyethylene. These LDPE polymers contain branched chains of polymerized monomer pendant from the main polymer "backbone" and generally have densities in the range of 0.910-0.935 gms/cc.

The other commercial technique involves coordination catalysts of the "Ziegler" or "Phillips" type and includes variations of the Ziegler type, such as the Natta type. These catalysts may be used at very high pressures, but generally are used at very low or

intermediate pressures. The products made by these coordination catalysts are generally known as "linear" polymers because of the substantial absence of branched chains of polymerized monomer pendant from the main polymer "backbone". They are also generally known as high density polyethylene (HDPE). Linear polyethylene (HDPE) ordinarily has a density in the range of 0.941 to 0.965 gms/cc.

10 The same coordination catalysts and polymerization technique are used in preparing copolymers of ethylene with other alpha-olefins. The effect of the comonomer is to lower the density below the HDPE range, but the polymer remains of the "linear" type.

The most preferred olefin polymers of the present invention are "linear" ethylene copolymers with minor amounts of alpha, beta-ethylenically unsaturated alkenes having from 3 to 12 carbons per alkene molecule, preferably 4 to 8. The amount of the alkene comonomer is generally sufficient to cause the density of the polymer to be about the same density range as LDPE, due to the alkyl sidechains on the polymer. Although the polymers remain in the "linear" classification, they are conveniently referred to as "linear low density polyethylene" (LLDPE). They retain much of the strength, crystallinity, and toughness normally found in HDPE homopolymers. Other polyolefins within the purview of the present invention, though less preferred than LLDPE, include HDPE, polypropylene, and polybutenes.

The present invention comprises a composition  
30 of an polyolefin resin and additives to form wettable  
fibers and fine filaments with high permanence of  
wettability. Since the neat polyolefins are hydrophobic

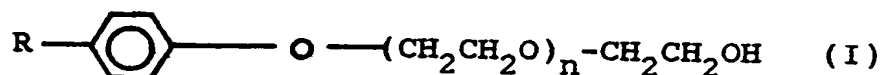
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materials, fibrous structures formed from polyolefin resins are not readily wet by water. In certain applications, such as those involving the dispersion of fibers in an aqueous medium and transport in or of an aqueous medium through an assembly of fibrous structures, this hydrophobic nature reduces the performance of polyolefin fibers. Imparting a lasting or use-variable surface wettability to polyolefin fibrous structures improves and expands their use as filtration structures, transport membranes and reinforcing matrices.

The polymer formulations of the present invention preferably contain 0.01 to 5 weight percent of the surface active (wetting) agent whether a single one, or a mixture of the agents. Most preferably 0.1 percent to 3 percent is used.

The surface active agent used in the practice of the present invention is at least one of the following: (1) an alkoxyated alkylphenol along with (or in combination with) a mixed mono-, di- and/or triglyceride, or (2) a polyoxyalkylene fatty acid ester, or (3) a combination of (2) and any or all parts of (1)

The alkoxyated alkylphenol preferably has the Formula I:

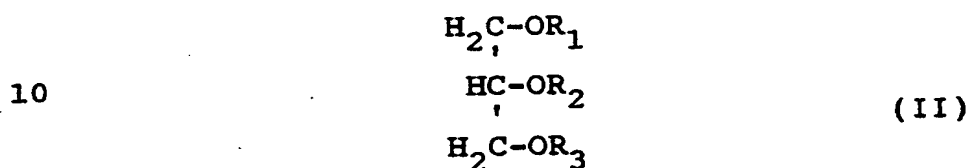


where R is an alkyl group of from 1 to 20 carbon atoms, preferably 5 to 15 carbon atoms, most preferably 6 to 12 carbon atoms; and where n is preferably a numerical value in the range of 10 to 55, more preferably in the

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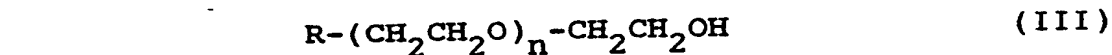
range of 10 to 30, most preferably in the range of 12 to 20. The numerical value n represents an average value as the length of the polyalkoxy chain may vary somewhat from molecule to molecule. A polyethoxy chain is the preferred polyalkoxy chain.

The mixed glyceride are exemplified by Formula II:



where  $\text{OR}_1$ ,  $\text{OR}_2$ , and  $\text{OR}_3$  represent independently hydroxyl or a fatty acid ester group, but at least one is a fatty acid ester. Thus, the mixed glyceride is a mono-, di-, or tri-glyceride of a fatty acid. The fatty acid may be saturated or unsaturated and is preferably a mixture of fatty acids having carbon chain lengths in the range of 12 to 18 carbons. Palm oil, e.g., is a convenient source of a mixture of fatty acids having carbon chain lengths within this range.

The preferred polyoxyalkylene fatty acid esters are exemplified by Formula III



where R is a fatty acid ester group, which may be saturated or unsaturated, and is preferably derived from a mixture of fatty acids having carbon chain lengths in the range of 12 to 18 carbons; and where n

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is preferably a numerical value in the range of 10 to 55, more preferably in the range of 10 to 30, most preferably 12 to 20. A polyoxyethylene chain is the preferred polyoxyalkylene chain.

5                   Mixing the surface active agents into the ethylene polymer is done with molten polymer and commonly used techniques such as rollmilling, mixing in a Banbury type mixer, or mixing in an extruder barrel. The heat history (time at which held at elevated temperature) can  
10 be shortened by mixing the surface active agent with unheated polymer particles so as to achieve substantially even distribution of the agent in the mass of polymer, thereby reducing the amount of time needed for intensive mixing at molten temperature.

15                   Conveniently, the surface active agent can also be added substantially simultaneously or sequentially with any other additives (colorants, dyes, etc.) which may be desired. The surface active agents may also be preblended with other additives and the blend  
20 then added to the polymer. In some instances these surface active agents have the additional benefit of aiding the other additives to become more easily or evenly dispersed or dissolved in the ethylene polymer. For easier batch-to-batch control of quality, it may  
25 be preferred to employ concentrated masterbatches of polymer/agent blends which are subsequently blended with additional quantities of polymer to achieve the final desired formulation. The masterbatch, or the neat additives, may be injected into freshly prepared  
30 polymer while the polymer is still molten after it leaves the polymerization vessel or train, and blended before the polymer is chilled to a solid or further processed.

Blends or alloys of olefin polymers, whether they be of the above described LDPE, LLDPE, HDPE or other olefin polymers or copolymers made using a free-radical initiator or a coordination catalyst may be used. Polypropylene is an example of an olefin polymer made using a coordination catalyst (e.g. the well-known Ziegler or Natta catalysts or variations thereof) which inherently exhibits a low density compared to polyethylene.

The use of polyolefin fibers is a growth area in the textile and related industries. Material advantages are being recognized as economic considerations drive the replacement of more expensive synthetic and natural fibers. The polyolefin fibers are making inroads in the disposable diaper market. Disposable diapers currently use a nonwoven, fibrous web as the skin contact innerliner. This innerliner should join the backing to hold the diaper together, transport fluid away from the skin via a wicking mechanism and provide a comfortable skin contact surface. The materials of choice for innerliners are presently polyester and cellulose with polypropylene gaining an increasing market share. The innerliners are composed of very fine, interconnected fibers of variable lengths. Polyester innerliners wet fairly readily and wick effectively but polyester webs have a coarse feel. Cellulose wets but also absorbs and retains water. Polypropylene provides a much softer web than polyester but it wets poorly, thus requires an added surface active agent. This limits the rate and efficiency of fluid transport due to the difficulty of forcing the fluid into the capillary matrix of the innerliners.



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Linear low density polyethylene (LLDPE) fibers exhibit enhanced tactile properties such as softness compared to polypropylene and high density polyethylene. Melt blending linear low resins with a combination of surface active agents and melt spinning the blend produces wettable fibers with tactile properties superior to fibers of barefoot (neat) linear low density polyethylene resins. Webs of the wettable linear low density polyethylene fibers exhibit rapid wetting and transport of aqueous media through the fiber matrix. These fibrous structures demonstrate good performance potential and offer a means for opening new markets for olefin polymers.

Wettability is a surface phenomena involving minimization of the interfacial surface energies between adjacent solid and liquid phases. In the case of water and polyolefins, wettability generally requires altering the polymer surface. This may be accomplished via a copolymer composition or by the action or auxilliary surfactants. Copolymers often detract from polyolefin material properties, add expense and make processing more difficult. Surface active agents are generally mobile species which aggregate as an interfacial compatibilizing layer on the polymer surface. The mobility of the surface layer makes it susceptible to solvation and mechanical dispersion. In other cases, where the surface active agents possess a strong affinity for the polyolefin substrate, fiber properties may deteriorate due to plasticization and/or detrimental structural rearrangements. Surfactants generally require an additional process step for application or activation and, in prior art, are often added after forming the fibrous or fabric product.

The present invention includes embodiments of a synergistic combination of surface active species, compounded directly with the resin, in contradistinction to post-added agents. The compound resin is fabricated by conventional processes and the wetting property is present in the product as fabricated. The resin in the present invention is easily processed and shows no detrimental change in properties. A broad range of wetting characteristics such as degree of wetting and permanence may be obtained by varying concentrations and composition of the additive package through the range of desired synergistic behavior. Resistance to solvation and mechanical dispersion is controlled by providing at least one surface active species that remains partially embedded in the substrate matrix while simultaneously participating in the interfacial zone.

This invention differs from the prior art by incorporating surface active agents directly into the bulk polymer resin rather than introducing a copolymer or applying a surface treatment to fabricated fibrous structures. The surface active agents bloom to the fabricated fiber surfaces where at least one of the surface active agents remain partially embedded in the polymer matrix. The permanence of wettability can be controlled through composition and concentration of the additive package. The more mobile components in the additive package can be designed to serve secondary functions such as skin conditioners or process aids.

The preferred blends of the present invention comprise about 95 percent to 99.9 percent of the olefin polymer, the remainder being the wetting agent or mixtures

The following examples further illustrate  
5 particular embodiments of the present invention.

An ethylene/1-octene copolymer with 2.5 MI and 0.935 g/cc density was melt blended with 1 percent by weight of a wetting agent package containing a combination of an ethoxylated alkylphenol with 14 units of ethylene oxide and a mixed glyceride with C<sub>12</sub>-C<sub>16</sub> fatty acid adduct (sold under the tradename Atmer 685). Fine filaments were spun at 250°C using an Instron capillary rheometer. The filaments were attenuated and collected with a variable speed roll. Wettability was compared to and examined along with filaments spun from the bare-foot (uncombined) resin using the same spinning conditions; four sets of each sample were tested.

Continuous filaments were wrapped around a metal support frame to form a flat, quasi-continuous surface. Droplets of deionized water were placed on the wrapped filament surfaces. The contact angles of the D.I. water droplets were measured on a Kayeness contact angle viewer with the results shown in Table I. The barefoot resin filament surface exhibited obtuse contact angles indicating a non-wetting surface. The resin plus additive filament surface wet immediately, resulting in contact angles less than  $1^\circ$ . The D.I. water on this surface passed through the adjacent filament interfaces.

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Wetting Test No. 2

Filaments from each resin sample were cut into short fibers and assembled into pseudo-nonwoven mats. D.I. water droplets were applied to the mat surfaces and visually observed without aid. Results are shown in Table I. Obtuse contact angles were again observed on the barefoot resin samples. Droplets remained on the surface until air dried. Sample mats made from the resin with the additive wetted instantly, allowing the water to migrate beneath the web structure. No surface water was visible.

Example 2

An ethylene/1-octene copolymer with 2.0 MI and 0.925 density was melt blended with 1 percent by weight of the same additive package used in Example 1. Fine filaments were spun at 220°C using the same equipment as in Example 1. Wettability was determined by Wetting Test No. 2. Permanence of wetting against abrasion was examined with two abrasion tests.

Abrasion Test No. 1

Rub small filament bundles twenty times between layers of a cellulosic laboratory wipe. Apply D.I. water droplets to the abraded bundle, time the penetration of water through the structure and note surface water.

Abrasion Test No. 2

Rub small filament bundles twenty times between closed fingers of a bare hand. Apply D.I. water droplets to the abraded bundle, time the penetration of the water into the structure and note surface water.

### Example 3

## 10 Example 4

### Example 5

### Example 6

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Example 7

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with 1 percent by weight of a wetting agent consisting of mono- and diglycerides (sold under the tradename of Atmos 300). Filaments were spun and tested as in Example 3 (see Table II under Example 7 for results).

Example 8

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with 1 percent by weight of a mono- and diglyceride mixture and 0.4 percent by weight of a nonylphenoxy poly(ethyleneoxy) ethanol (9 moles ethylene oxide). Filaments were spun and tested as in Example 3 (see Table II under Example 8 for results).

Example 9

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with 1 percent by weight of a wetting agent, polyoxyethylene laurate (sold by C. P. Hall Chemical Company under the trade-name CPH 376-N). Filaments were spun and tested as in Example 3 (see Table II under Example 9 for results).

Example 10

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with 1 percent by weight of a wetting agent, nonylphenoxy poly(ethyleneoxy) ethanol, 20 moles ethylene oxide. Filaments were spun and tested as in Example 3 (see Table II under Example 10 for results).

Example 11

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with 1 percent by weight of a wetting agent, nonylphenoxypoly(ethylene-oxy)ethanol (30 moles ethylene oxide). Filaments were spun and tested as in Example 3 (see Table II for results, under Example 11).

Example 12

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with an additive package to give 0.5 percent by weight of mono- and diglycerides and 0.5 percent by weight of nonylphenoxypoly(ethyleneoxy)ethanol (20 moles ethylene oxide). Filaments were spun and tested as in Example 3 (see Table II for results, under Example 12).

Example 13

An ethylene/1-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with an additive package to give 0.5 percent by weight of mono- and diglycerides and 0.5 percent by weight of nonylphenoxypoly(ethyleneoxy)ethanol (30 moles ethylene oxide). Filaments were spun and tested as in Example 3 (see Table II for results, under Example 13).

Example 14

An ethylene 1/-octene copolymer with 2.3 MI and 0.917 g/cc density was melt blended with 1 percent by weight of an ethoxylated mono- and diglyceride and sold under the tradename Sherex LI-42. Filaments were spun and tested as in Example 3 (see results in Table II, under Example 14).

Example 15

The filaments from the blended resin of Example 3 were soaked in 1N HCl and 1 percent  $\text{NH}_4\text{OH}$  for 24 hours. The filaments were then rinsed thoroughly in D.I.  $\text{H}_2\text{O}$  and air dried overnight. The filaments wetted completely within one second when droplets of D.I. water were applied to the bundle surface.

Example 16

An ethylene/1-octene copolymer with 6.0 MI and 0.919 g/cc density was melt blended with 1 percent by weight of the additive package of Example 1. A continuous filament bundle was produced using a screw extruder to feed a melt gear pump and spinnerette pack. The filament bundle was air quenched and collected by mechanical wind up at 1000-1500 m/min. Also some of the filament bundle was collected by air laydown into a screen in random web. The as-spun filaments showed instant wetting by water as judged by Wetting Test No. 2. 2.5 cm (1 in) staple fibers, cut from these filaments, were processed into nonwoven webs or a card, a Garnett, and a Rando-Webber. These nonwoven webs exhibited instant aqueous wetting as judged by Wetting Test No. 2.



## Wetting Test

### Test No.2

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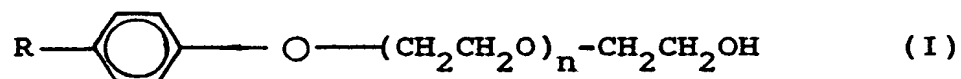
TABLE II

Example No.	Wetting Test No. 2	Abrasion Test No. 1	Abrasion Test No. 2
2	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
3	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
4	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
5	Wet Slowly; >1 min.	<min., Completely	<10 sec., Completely
6	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
7	<1 sec., Surface Layer	<3 sec., Surface Layer	<5 sec., Surface Layer
8	<2 sec., Surface Layer	~5 sec., Surface Layer	~5 sec., Surface Layer
9	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
10	>10 sec., Surface Layer	>2 min., No Wetting	>2 min., No Wetting
11	No Wetting	>2 min., No Wetting	>2 min., No Wetting
12	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
13	Sorbed, Wet Instantly	Wet Instantly, Completely	Wet Instantly, Completely
14	No Wetting	>2 min., No Wetting	>2 min., No Wetting

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1. Wettable fibers or fine filaments prepared from a composition comprising an olefin polymer having incorporated therein a wetting agent, said wetting agent comprising at least one of (1) an alkoxyated alkylphenol along with, or in combination with, a mixed mono-, di- and/or triglyceride, (2) a polyoxyalkylene fatty acid ester, or (3) a combination of (2) and any or all parts of (1) and said olefin polymer is a linear low density polyethylene (LLDPE) and blends or alloys of LLDPE with other olefin polymers.

2. The fibers or fine filaments of Claim 1 wherein (A) the alkoxyated alkylphenol has Formula I:

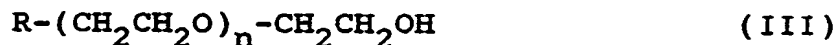


where R is an alkyl group of from 1 to 20 carbon atoms, and n is a numerical value in the range of 10 to 55;  
(B) the mixed glyceride has Formula II:



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where  $OR_1$ ,  $OR_2$ , and  $OR_3$  represent, independently hydroxyl or a fatty acid ester group, but at least one is a fatty acid ester; and (C) the polyoxyalkylene fatty acid ester has Formula III:



where R is a fatty acid ester group, either saturated or unsaturated, and n is a numerical value in the range of 10 to 55.

3. The fibers or fine filaments of Claim 1 wherein the olefin polymer is linear low density polyethylene (LLDPE).

4. The fibers or fine filaments of Claim 1 wherein the olefin polymer is LLDPE blended or alloyed with low density polyethylene (LDPE), high density polyethylene (HDPE), polypropylene and/or polybutene.

5. The fibers or fine filaments of Claim 1 wherein the olefin polymer is a copolymer of ethylene and at least one  $C_3$ - $C_{12}$  alpha-olefin.

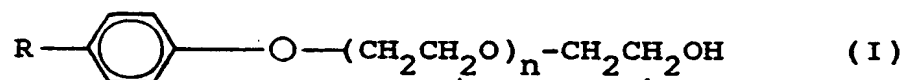
6. The fibers or fine filaments composition of Claim 1 wherein the wetting agent is present in an amount of 0.01 percent to 5 percent by weight.

7. The fibers or fine filaments of Claim 1 wherein the fibers or filaments comprise a woven fabric, a non-woven fabric, or a knitted fabric.

8. Wettable fibers or fine filaments prepared from a composition comprising an olefin polymer having incorporated therein a wetting agent, said wetting agent comprising at least one combination of:

- (a) an alkoxyated alkyl phenol along with a mixed mono-, di-, and/or triglyceride;
- (b) an alkoxyated alkyl phenol along with a polyoxyalkylene fatty acid ester;
- (c) a polyoxyalkylene fatty acid ester along with a mixed mono-, di- and/or triglyceride; or
- (d) an alkoxyated alkyl phenol along with a mixed mono-, di-, and/or triglyceride and with a polyoxyalkylene fatty acid ester.

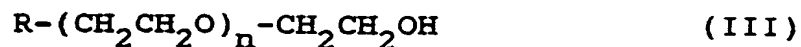
9. The wettable fibers or fine filaments of Claim 25 wherein (a) the alkoxyatedalkyl phenol has Formula I



where R is an alkyl group of from 1 to 20 carbon atoms, and n is a numerical value in the range of 10 to 55; (b) the mixed glyceride has Formula II.



where  $\text{OR}_1$ ,  $\text{OR}_2$ , and  $\text{OR}_3$  represent, independently hydroxyl or a fatty acid ester group, but at least one is a fatty acid ester; and (c) the polyoxyalkylene fatty acid ester has Formula III:



where R is a fatty acid ester group, either saturated or unsaturated, and n is a numerical value in the range of 10 to 55.

10. The wettable fibers or fine filaments of Claim 9 when employed as a wettable portion of products of the group comprising diaper products, battery cell separators, filters, papers, membranes, diaphragms, and construction materials.

11. The wettable fibers or fine filaments of Claim 9 in the form of a dispersion in an aqueous medium.